

CLAIMS

1. A multi-layer film having a layer structure that a polyvalent metal compound-containing layer (B) adjoins one side or both sides of a polymer layer (A) containing a carboxyl group-containing polymer and a polyvalent metal salt of the carboxyl group-containing polymer, wherein

(1) the polymer layer (A) has a concentration gradient structure that the concentration of the polyvalent metal salt of the carboxyl group-containing polymer in the polymer layer (A) is continuously reduced in the thickness-wise direction from a surface adjoining the polyvalent metal compound-containing layer (B), and

(2) a peak ratio A_{1560}/A_{1700} of the height A_{1560} of an absorption peak at a wave number of 1560 cm^{-1} to the height A_{1700} of an absorption peak at a wave number of 1700 cm^{-1} as determined on the basis of an infrared absorption spectrum of the polymer layer (A) is at least 0.25.

2. The multi-layer film according to claim 1, wherein the multi-layer film has a layer structure of the polymer layer (A)/the polyvalent metal compound-containing layer (B), and the polymer layer (A) has a concentration gradient structure that the concentration of the polyvalent metal salt of the carboxyl group-containing polymer in the polymer layer (A) is continuously reduced up to a surface opposite to a surface adjoining the polyvalent metal

compound-containing layer (B) of the polymer layer (A) in the thickness-wise direction from the adjoining surface.

3. The multi-layer film according to claim 1,
5 wherein the multi-layer film has a layer structure of the polymer layer (A)/the polyvalent metal compound-containing layer (B)/the polymer layer (A), and each of the polymer layers (A) has a concentration gradient structure that the concentration of the polyvalent metal salt of the carboxyl
10 group-containing polymer in the polymer layer (A) is continuously reduced up to a surface opposite to a surface adjoining the polyvalent metal compound-containing layer (B) of the polymer layer (A) in the thickness-wise direction from the adjoining surface.

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4. The multi-layer film according to claim 1,
wherein the multi-layer film has a layer structure of the polyvalent metal compound-containing layer (B)/the polymer layer (A)/the polyvalent metal compound-containing layer
20 (B), and the polymer layer (A) has a concentration gradient structure that the concentration of the polyvalent metal salt of the carboxyl group-containing polymer in the polymer layer (A) is continuously reduced up to a central portion of the polymer layer (A) in the thickness-wise
25 direction from a surface adjoining each of the polyvalent metal compound-containing layers (B).

5. The multi-layer film according to claim 1,
wherein the concentration gradient structure of the polymer
layer (A) has a low concentration region of the polyvalent
metal salt of the carboxyl group-containing polymer, whose
5 element mole number ratio $[\text{metal element (n valence)}/\text{C}$
element] calculated out on the basis of the result of
elemental composition analysis in the thickness-wise
direction by energy dispersive X-ray spectroscopy falls
within a range of 0 to $0.06/n$, in a proportion of 5 to 80%
10 in terms of a thickness percentage of the polymer layer (A).

6. The multi-layer film according to claim 1,
wherein the concentration gradient structure of the polymer
layer (A) has a high concentration region of the polyvalent
15 metal salt of the carboxyl group-containing polymer, whose
element mole number ratio $[\text{metal element (n valence)}/\text{C}$
element] calculated out on the basis of the result of
elemental composition analysis in the thickness-wise
direction by energy dispersive X-ray spectroscopy falls
20 within a range of from higher than $0.06/n$ to not higher
than $0.33/n$, in a proportion of 20 to 95% in terms of a
thickness percentage of the polymer layer (A) adjoining the
polyvalent metal compound-containing layer (B).

25 7. The multi-layer film according to claim 2,
wherein the polymer layer (A) has a high concentration
region of the polyvalent metal salt of the carboxyl group-

containing polymer, whose element mole number ratio [metal element (n valence)/C element] calculated out on the basis of the result of elemental composition analysis in the thickness-wise direction by energy dispersive X-ray spectroscopy falls within a range of from higher than 0.06/n to not higher than 0.33/n, in a proportion of 20 to 95% in terms of a thickness percentage of the polymer layer (A) adjoining the polyvalent metal compound-containing layer (B), and has, on the side opposite to the surface adjoining the polyvalent metal compound-containing layer (B), a low concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] falls within a range of 0 to 0.06/n, in a proportion of 5 to 80% in terms of a thickness percentage of the polymer layer (A).

8. The multi-layer film according to claim 3, wherein each of the polymer layers (A) has a high concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] calculated out on the basis of the result of elemental composition analysis in the thickness-wise direction by energy dispersive X-ray spectroscopy falls within a range of from higher than 0.06/n to not higher than 0.33/n, in a proportion of 20 to 95% in terms of a thickness percentage

of the polymer layer (A) adjoining the polyvalent metal compound-containing layer (B), and has, on the side opposite to the surface adjoining the polyvalent metal compound-containing layer (B), a low concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] falls within a range of 0 to $0.06/n$, in a proportion of 5 to 80% in terms of a thickness percentage of the polymer layer (A).

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9. The multi-layer film according to claim 4, wherein the polymer layer (A) has a high concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] calculated out on the basis of the result of elemental composition analysis in the thickness-wise direction by energy dispersive X-ray spectroscopy falls within a range of from higher than $0.06/n$ to not higher than $0.33/n$, in a proportion of 20 to 95% in terms of a thickness percentage of the polymer layer (A) adjoining each of the polyvalent metal compound-containing layers (B), and has, at a central portion thereof, a low concentration region of the polyvalent metal salt of the carboxyl group-containing polymer, whose element mole number ratio [metal element (n valence)/C element] falls within a range of 0 to $0.06/n$, in a proportion of 5 to 80% in terms of a thickness percentage

of the polymer layer (A).

10. The multi-layer film according to claim 1,
wherein a chemical equivalent of the polyvalent metal
5 compound to the carboxyl group, which is calculated out on
the basis of the total (At) of the carboxyl group contained
in the whole carboxyl group-containing polymer layer (A)
and the total (Bt) of the polyvalent metal compound
contained in the whole carboxyl group-containing polymer
10 layer (A) and the whole polyvalent metal compound-
containing layer (B), is at least 1.0.

11. The multi-layer film according to claim 1,
wherein an oxygen transmission rate as determined by using
15 the multi-layer film formed into a cylindrical form as a
sample to conduct a flex test of 150 Gelvo flexings by
means of a Gelvo tester under conditions of a temperature
of 5°C and a relative humidity of 10% in accordance with
the provisions of ASTM F 392, aging the sample for 20 hours
20 under an environment of a temperature of 30°C and a
relative humidity of 80%, and then measuring an oxygen
transmission rate of the sample after the aging under
conditions of a temperature of 30°C and a relative humidity
of 0% in accordance with ASTM D 3985-81 is retained within
25 $\pm 50\%$ of the oxygen transmission rate of the sample before
the flex test.

12. The multi-layer film according to claim 1,
wherein the carboxyl group-containing polymer has an oxygen
transmission coefficient of at most $1,000 \text{ cm}^3 \cdot \mu\text{m}/$
 $(\text{m}^2 \cdot \text{day} \cdot \text{MPa})$ as determined in the form of a film formed by
5 itself under low-humidity conditions of a temperature of
30°C and a relative humidity of 0%.

13. The multi-layer film according to claim 1,
10 wherein the carboxyl group-containing polymer is a
homopolymer of a carboxyl group-containing unsaturated
monomer, a copolymer of carboxyl group-containing
unsaturated monomers, a copolymer of a carboxyl group-
containing unsaturated monomer and any other polymerizable
15 monomer, a carboxyl group-containing polysaccharide or a
mixture of at least two monomers thereof.

14. The multi-layer film according to claim 1,
wherein the carboxyl group-containing unsaturated monomer
20 is at least one α, β -monoethylenically unsaturated
carboxylic acid selected from the group consisting of
acrylic acid, methacrylic acid, crotonic acid, itaconic
acid, maleic acid and fumaric acid.

25 15. The multi-layer film according to claim 1,
wherein the polyvalent metal compound is an oxide,
carbonate, organic acid salt or alkoxide of beryllium,

magnesium, calcium, copper, cobalt, nickel, zinc, aluminum or zirconium.

16. The multi-layer film according to claim 1,
5 wherein the polyvalent metal compound is a divalent metal compound.

17. The multi-layer film according to claim 1,
wherein the thickness of the polymer layer (A) is 0.001 μm
10 to 1 mm, and the thickness of the polyvalent metal compound-containing layer (B) is 0.001 μm to 1 mm.

18. The multi-layer film according to claim 1, which is obtained by a process comprising forming at least one
15 carboxyl group-containing polymer layer and at least one polyvalent metal compound-containing layer adjacently to each other on the base by a coating method and then aging the polyvalent metal compound-containing layer under an atmosphere of a relative humidity of at least 20% and a
20 temperature of 5°C to 200°C to cause the polyvalent metal compound to migrate from the polyvalent metal compound-containing layer into the carboxyl group-containing polymer layer, thereby forming a polyvalent metal salt with the carboxyl group in the carboxyl group-containing polymer
25 layer.

19. The multi-layer film according to claim 1, which

has an oxygen transmission rate of at most $1,000 \text{ cm}^3(\text{STP}) / (\text{m}^2 \cdot \text{day} \cdot \text{MPa})$ as determined under high-humidity conditions of a temperature of 30°C and a relative humidity of 80%.

- 5 20. A multi-layer film obtained by additionally arranging at least one another resin layer on one side or both sides of the multi-layer film according to any one of claims 1 to 19.